Roll to Roll (R2R) Processing Technology Assessment

Contents

1. Introduction to the Technology/System

1.1. Introduction to R2R Processing

Roll-to-roll (R2R) is a family of manufacturing techniques involving continuous processing of a flexible substrate as it is transferred between two moving rolls of material [1]. R2R is an important class of substrate-based manufacturing processes in which additive and subtractive processes can be used to build structures in a continuous manner. Other methods include sheet to sheet, sheets on shuttle, and roll to sheet; much of the technology potential described in this R2R Technology Assessment conveys to these associated, substrate-based manufacturing methods [2]. R2R is a "process" comprising many technologies that, when combined, can produce rolls of finished material in an efficient and cost effective manner with the benefits of high production rates and in mass quantities. High throughput and low cost are the factors that differentiate R2R manufacturing from conventional manufacturing which is slower and higher cost due to the multiple steps involved, for instance, in batch processing. Initial capital costs can be high to set up such a system; however, these costs can often be recovered through economy of scale. Figure 1 illustrates an example of R2R processing of a state-of-the-art nanomaterial used in flexible touchscreen displays. [3]

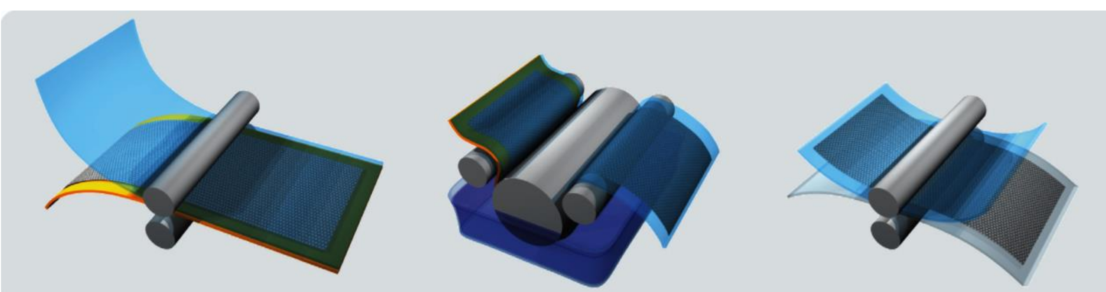

Figure 1 – R2R processing of graphene film for flexible touchscreen displays [3].

Today, R2R processing is applied in numerous manufacturing fields such as flexible and large-area electronics devices, flexible solar panels, printed/flexible thin-film batteries, fibers and textiles, metal foil and sheet manufacturing, medical products, energy products in buildings, and membranes to name a few. In the field of electronic devices, R2R processing is a method of producing flexible and large-area electronic devices on a roll of plastic or metal foil. Substrate materials used in R2R printing are typically paper, plastic films or metal foils. Stainless steel is sometimes used because it is durable and has a high temperature tolerance [4]. The global flexible electronics (flexible display, flexible battery, flexible sensor, flexible memory and thin film photovoltaic (PV)) market revenue was estimated to grow from $3.4 billion in 2013 to $13.23 billion in 2020 at a compound annual growth rate (CAGR) of 21.73% from 2014 to 2020. The consumer electronics market is expected to grow at a CAGR of 44.30% and is supported by advancements in flexible displays, flexible sensors and thin-film solid-state batteries that can be produced using R2R processes. [1] [5]

Further development of R2R production capabilities that are energy efficient, low environmental impact and lower cost and that are employed to manufacture technologies and products for clean energy applications will have a "global impact" in the manufacturing industry. There are huge savings in energy just from higher throughputs since the tools and equipment used in R2R manufacturing (per unit area of manufactured roll) are using less energy for a much shorter period of time relative to conventional manufacturing processes. Additionally, efficiencies are obtained from more efficient deposition processes, for example, that would provide additional savings in energy. Breakthroughs that will have high impact, and therefore high value, are in the nano-manufacturing community. [1]

The R2R Tech Assessment reviews current state-of-the-art technologies, clean energy applications, and industry investments to categorize advances in R2R manufacturing in the areas of metrology, equipment, carriers/webs, substrate materials, process improvement, alternative applications and other possible innovations. These efforts will serve to enable and maintain the competitive nature of R2R manufacturing for the domestic U.S. industry.

1.2. R2R Processing Mechanisms

Silicon wafers, cadmium-telluride solar cells, battery electrodes, fuel cell membranes, and high performance window films are just a few examples of materials that have clean energy applications and are characterized by a two-dimensional functional surface, often with one or more coated or deposited layers. Not surprisingly, these materials are often made using similar processes—namely continuous roll-to-roll, belt-fed, or conveyor-based processes that enable successive steps to build a final construction at high throughput.

As a comparison of the variety of processes that can be used for R2R manufacturing, a brief description of each are provided here.

- **Deposition** – Evaporation, sputtering, and chemical vapor deposition (CVD) can all be easily implemented in R2R processing. Multilayer sputtering systems are the most common. The entire roll is loaded into a vacuum system where it is relatively easy to sputter or evaporate different materials onto a substrate without crosstalk as shown in Figure 2. This is more difficult in CVD where reactive gas barriers are needed within the vacuum system. [7] When the substrate moves past the sputtering source, the deposition rate of material varies. The processing rate influences the thickness and sequence of layers in a multilayer coating which also depends on rotation speed, initial position and orientation of the substrate. CVD can be used to deposit materials on a continuous roll of flexible metal foils, plastics, and other materials in place of individual substrates. This technology has been used for superconductor tape production and nanomaterial synthesis and is growing in popularity for thin film solar deposition.

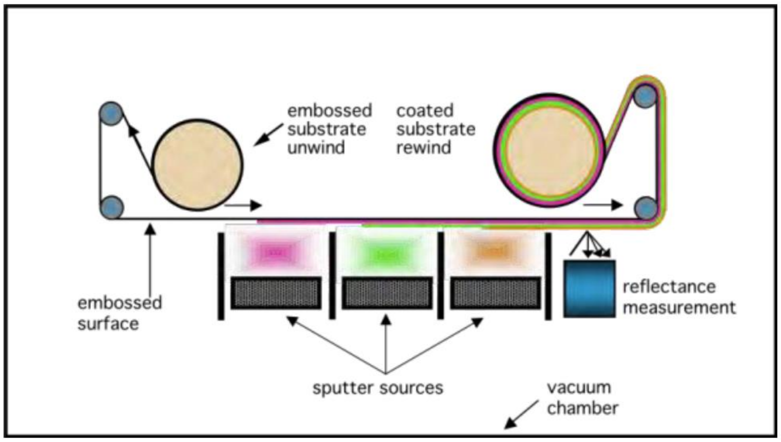

Figure 2 – Inline vacuum coater and sputtering process [1].

- **Gravure** - A type of printing process which involves engraving the image onto an image carrier. In gravure printing, the image is engraved onto a cylinder because, like offset printing and flexography, it uses a rotary printing press. The entire patterned cylinder is covered with ink as shown in the upper left corner of Figure 3. The excess ink is doctored off, leaving ink in the cup-shaped engraved pattern. The plate cylinder is brought into contact with the impression cylinder to transfer the ink to the substrate. [8] Once a staple of newspaper photo features, the process is still used for commercial printing of magazines, postcards, and product packaging.

- **Flexographic Printing** - A form of printing process which utilizes a flexible relief plate as shown in the upper right corner of Figure 3. It is essentially a modern version of letterpress which can be used for printing on almost any type of substrate, including plastic, metallic films, cellophane, and paper. It is widely used for printing on the non-porous substrates required for various types of food packaging. Only the raised area in the pattern cylinder is inked and the pattern is transferred to the substrate. [8]

- **Flatbed and Rotary Screen Printing** – In flatbed printing, a squeegee, moves relative to a mesh, then forces the ink through the open area and onto the substrate. The wet layer thickness is defined by the thickness as well as the open area of the mesh and generally relative thick wet layers can be achieved (10–500 µm). [9] In rotary screen printing, the substrate moves through rollers past the squeegee forcing the ink onto the substrate. Both of these processes are illustrated in the lower half of Figure 3.

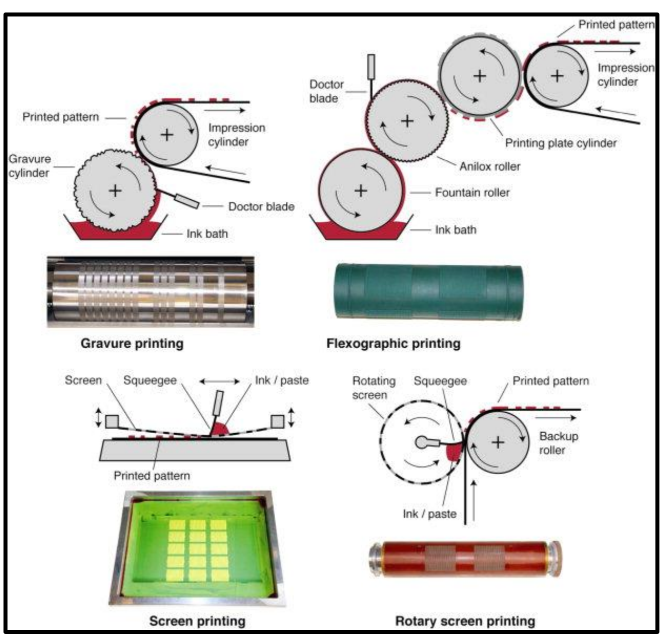

Figure 3 - R2R Processing diagrams for organic electronics/thin films [6].

- **Imprint or Soft Lithography** – In soft lithography (e.g. self-aligned imprint lithography (SAIL)), multiple mask levels are imprinted as a single three dimensional (3-D) structure as shown in Figure 4. The photopolymer layer is heated above its glass transition temperature to allow it to flow into the crevices of the stamp. The stamp/ polymer sandwich is cured with ultraviolet (UV) light as the polymer cools and hardens, allowing the stamp to pull off cleanly. The process is completed with standard wet and dry etch processes, leaving an accurately reproduced 3-D, high resolution pattern on the substrate. The technology is called self-aligning because the mask would deform with the substrate during the embossing heat treatment step. [10]

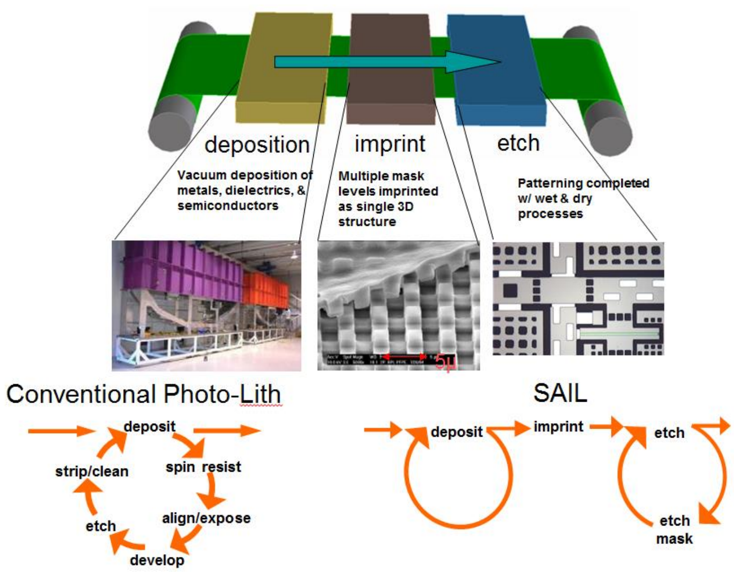

Figure 4 – Self-Aligned Imprint Lithography [11].

- **Laser Ablation** - A technique that would eliminate both the photoresist coating and wet etching steps is called laser photoablation and is illustrated in Figure 5. This technique is used to write directly into a polymer layer using a high powered laser. The photoablation works by breaking molecular bonds in polymer layer, fracturing the polymer into shorter units that are “kinetically ejected" upon removal. The amount of material ejected can be tuned by adjusting the wavelength, energy density and pulse width of the xenon-flouride (XeF) excimer laser (a form of UV laser which is commonly used in the production of microelectronic devices) used for ablation and is capable of reproducing ablation depth to within 0.1 µm across large areas of the substrate. Examples of ablatable polymers are polyimides (e.g "Kapton®") and polyethylene terephthalate (PET) (e.g Mylar®), which are also commonly used substrates in flexible electronics [10].

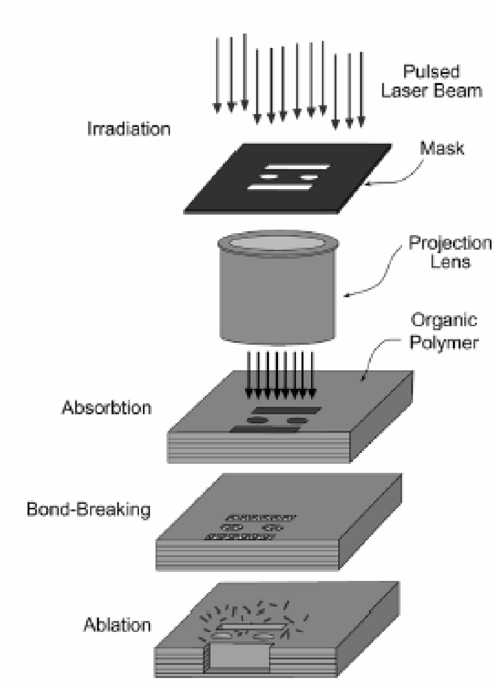

Figure 5 – Schematic of the laser ablation process [10].

- **Offset Printing** - A commonly used technique in which the inked image is transferred (or "offset") from a blanket cylinder that bridges the plate cylinder and the substrate. The pattern is transferred to the blanket (usually made of rubber), and then transferred to the substrate [8].

- **Inkjet Printing** - While laser ablation may be called a subtractive technique, inkjet printing can be considered an additive technique. Rather than your home, graphics-oriented inkjet printer, an array of piezoelectric print heads are required for the deposition of conducting organic solutions at precise locations [10].

Table 1 provides a comparison between some of these different printing methods in terms of their theoretical capacity and practical applicability for large-scale R2R production.

Table 1 – Comparison between different printing methods in terms of their theoretical capacity and practical applicability for large-scale R2R production

Printing Method	Speed	Wet Thickness (µm)	Resolution (µm)	Start/Stop	Complexity	Applicability
Flatbed Screen Printing [5]	Low	5-100	100	Yes	Low	Limited
Rotary Screen Printing [5]	High	3-500	100	Yes(a)	Medium	Very good
Inkjet Printing [5]	Medium	1-5	<50	Yes	High	Limited, materials must be jettable
Flexography [5]	Very high	1-10	<50	Yes(a)	Medium	Very good

Imprint or soft lithography [20]	High (>5 meters/min)		0.1 (100 nm demonstrated)			New technology
Laser ablation [20]	Low		~10			Thermal effect sensitivity
Gravure [12]	High		>0.07 (70 nm demonstrated)			Very Good

(a) - Stopping should be avoided. Risk of registration lost and drying of ink in anilox cylinder. Short run-in length

Substrates that are used in R2R processing may be made of a variety of materials depending on the application and processing steps involved in fabrication. Plastic films are desirable for their transparency, flexibility and toughness, but are often susceptible to degradation and dimensional distortion at high temperature [10]. Where transparency is not required, stainless steel foils may be chosen as they tolerate higher temperatures than plastics. Other materials, such as aluminum and copper alloys, may also be used.

Circuit patterns may be formed on the flexible substrate in a variety of ways. Some such production lines employ inkjet technology to deposit material onto the substrate. This process is similar to how an inkjet printer deposits ink onto paper. Some facilities employ photolithography, using light to etch away a pattern on the substrate that may then be filled with another material. Other techniques using ultraviolet light, lasers, and so on, may also be used to imprint the substrate with electrical circuitry.

A variety of other manufacturing steps may also be accomplished using R2R processing. In addition to laying down circuit patterns, such steps as die cutting, laminating, placing labels, cleaning, and more may be performed. Heat sealing and application of a variety of coatings may also be included in a roll-to-roll processing operation.

2. Technology Assessment and Potential

2.1. Benefits of R2R Manufacturing

Benefits of R2R processing include high production rates and yields. This technique can help reduce the cost of manufacturing through economy of scale as it allows devices to be fabricated automatically in mass quantities. Although initial capital costs can be high to set up such a system, these costs can often be recovered through the economic advantages during production [13]. For conventional sheet-fed systems, sheet handling and off-line drying consume a good portion of the overall cycle time. Continuous production can be achieved on a roll-to-roll system due to in-line hot air drying and sophisticated web-tension controls.

2.2. R2R Processing Applications

The R2R technology has evolved to support a wide range of industrial applications used for both traditional and “cutting-edge” products [1]. Two flexible thin film products made by R2R processing are shown in Figure 6.

Figure 6 – Example battery electrode (left) and thin film (right) photovoltaic materials. Photo from David Wood, Oak Ridge National Laboratory (L) and Warren Gretz, NREL

The major technology areas with clean energy applications that have been produced using R2R manufacturing are as follows:

- Flexible electronics - super-capacitors, electronic circuits, radio frequency identification (RFID) chips, organic light emitting diodes (OLEDs), displays, sensors, etc.
- Flexible photovoltaics - Copper-Indium Gallium-Selenide Photovoltaic (CIGS PV) and other flexible PV products (Figure 6),
- Printed/flexible thin-film batteries - laminar Lithium-ion, etc. (Figure 6),
- Fuel cells - laminar solid oxide fuel cells (SOFC), proton exchange membranes (PEM), membrane electrode assemblies and gas diffusion media,
- Multilayer capacitors (MLC), (i.e. dielectrics such as NPO/X5R/XR7/Relaxer/etc.)
- Thick and thin-film substrates (Al_2O_3, AlN, Si_3N_4, SiC, GaN, MgO, ZrO, etc.)
- Thick-film sensor materials (temperature sensors, positioners, transducers, e.g. negative temperature coefficient thermistors, piezoelectric/lead zirconate titanate (PZT), active/passive, selective gas)
- Fabric (clothing textiles, fiber reinforce mat/fiberglass/carbon/polymer)
- Anti-static, release, reflective and anti-reflective coatings (glass, Mylar®, polyethylene)
- Barrier Coatings (thermal and environmental)
- Building products, films (electro-chromic, reflectives, etc.), composite structural members, etc.
- Metal ribbon (transformer “coils”, etc.)
- Paper industry
- Chemical separation membranes (reverse osmosis, catalyst)

All are considered to be 2-D processed using continuous sheet-based manufacturing lines which have been developed in a variety of forms. Figure 7 shows two generic R2R processes of screen printing and tape casting. R2R lines are used when a continuous sheet, or “web”, can be conveyed on the line in an unsupported fashion. In addition to the web speed, the tension of the web is typically controlled to ensure that the motion of the web across and around a multiplicity of rollers is done in a way that does not cause stretching or wrinkling of the web. Belt-fed lines are similar and are used when support of the web during processing is required, for example during high-temperature process steps. Float lines are similar in concept and allow long sheets of material such as glass to be processed while moving and supported on a liquid surface. Finally, conveyors are used for cases such as silicon photovoltaic wafers wherein discrete parts are processed in a continuous fashion, although much processing is accomplished using batch process methods.

Figure 7 - (Left) screen printing and (right) tape casting.
Photos from M. Richards, Versa Power Systems

Many different permutations of processes are used on these continuous lines—too many to review in detail. Instead, broad categories of processes are highlighted below. A high level discussion on various printing/coating/deposition mechanisms was already covered in section 1.2. Most of the materials of interest involve some kind of coating or deposition—often several in series—to create functional layers and surfaces [7]. These additive processes are categorized by the pressure at which the coating is applied: either at atmospheric (room) pressure or in a vacuum. Atmospheric coatings take several generic forms. Roll coating is characterized by two or more rollers, in a wide variety of configurations, being used to "pick up" a thin layer of liquid from a bath and apply it to a surface of a web. Knife coating is similar to roll coating, wherein a stationary bar or rod—the "knife"—, commonly known as a "Dr. Blade" is set to a certain stand-off distance from the web and is used to control the amount of liquid deposited onto the web from a reservoir in process referred to as tape casting [14]. Figure 8 illustrates this process.

Figure 8 - Traditional, current technology "Laboratory-Scale" Tape Caster and "Dr. Blade", manufactured by HED used to deposit thick-film slurry on moving web "substrate.

Various masks or other limits to the location or position of the coated liquid can be employed, as in screen printing. A wide variety of techniques are generically referred to as die coating, characterized by a sheet of coating being dropped or laid onto the web. The die comprises two or more typically metal plates with machined flow-fields between to enable the creation of a highly uniform sheet of coating. And finally, for the atmospheric coatings, spray methods are often employed using one or an array of spray heads to coat the web from side to side. Low temperature systems are used most often, including a variety of jet methods as well as systems where the head is ultrasonically actuated to break up droplets and particles into a very fine spray. In cases where the substrate or base material can withstand the thermal load, high temperature sprays can be used, including electrical arc and plasma-based methods. In almost all cases of liquid coatings applied under atmospheric pressure, some kind of drying and/or curing of the coating is required. Drying is used to drive off solvents that are used to make a coatable mixture but are not desired in the final layer and is typically accomplished using heated gas or infrared heat sources. Curing is a post-treatment process to finalize the chemical or morphological nature of the coating by irradiation with an energy source such as infrared or ultraviolet lamps, or an electron beam.

Vacuum coating techniques incorporate a number of vapor deposition technologies, such as sputtering and evaporative coating. These processes are typically used for very thin coatings—usually less than a micrometer in thickness, referred to thin-film processes [15]. Importantly, when vacuum processes are used in continuous production, complicated and expensive line equipment must be employed to allow movement of the web while still maintaining very low pressure. Several mechanical processing steps are also used including cutting or sawing processes, texturing of the surface, and creation of electrical junctions. Figure 9 shows a reel-to-reel vacuum deposition process.

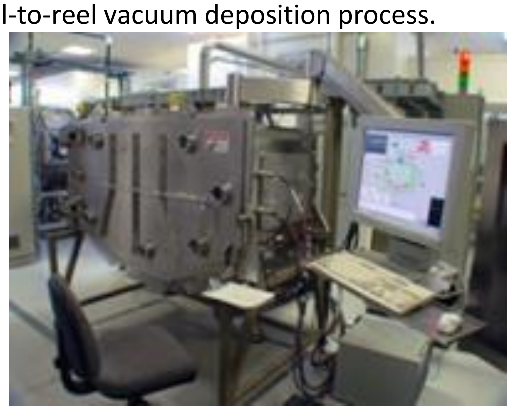

Figure 9 - Reel-to-reel vacuum deposition line.
Photo from Global Solar Energy, NREL 13414

Many of the described processes have been available to the manufacturer for many, i.e. > 40 years. An idealized R2R manufacturing process with the essential steps from the raw materials to the finished product is illustrated in Figure 10. However, because of demand for increased process competitiveness, new applications and equipment, researchers have continued to evolve the wide range of processes to meet innovation challenges. Whereas in the 1970s one was barely able to print 25 µm wide lines and traces on a 250 µm thick substrate, today, it has been demonstrated that investigators can routinely printi sub-200 nm features in a continuous web [16].

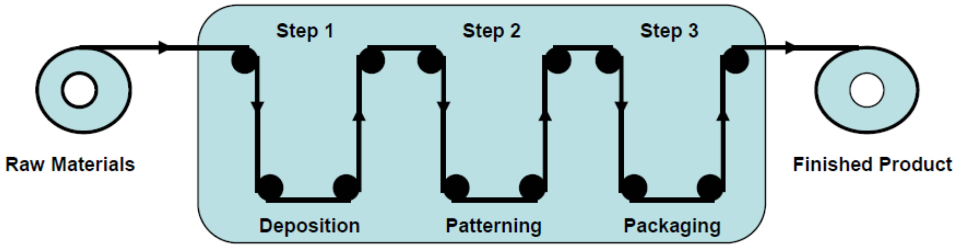

Figure 10 - Idealized Roll-to-Roll manufacturing process flow [4]

Regardless of the process, each technology where 2-D processing is used exhibits a wide range of net process yields. In both the thick-film and thin-film microelectronics industries substrate surface imperfections, chemical impurities inconsistencies in substrate thickness, flatness and planarity can all result in considerable yield loss. These losses do not include additional issues due to improper deposition of conductors, devices, vias (small opening for an electrical connection) and other through-holes, mask alignments, etch issues, oxidation, etc. Nor do these losses include "pre-fabrication" yield losses during ingot preparation, doping, thermal processing, wafering, dicing and other more mechanical issues as simple as "breakage". Overall "net-yields" for thick-film transducer and thermistor products were as low as 25 to 30 percent, while those encountered for select discrete thin-film fabricated products ranged to lows of 40 to 60 percent. Indeed 2 dimensional R2R processing is one which provides a manufacturer with a means to rapidly increase capacity and productivity. However, the specific process in-use and all tools, materials, designs, etc., must be reliable, reproducible and be capable of consistently yielding product to the prerequisite specifications. The "openness" of government and industry to form partnerships will solve challenges with substrate materials, morphology, and hardware/software that are common in all applications. [4]

2.3. Challenges to R2R Manufacturing

The following summarizes some of the challenges faced by industry when considering R2R manufacturing to produce a technology.

- In order to succeed as a viable manufacturing alternative, R2R processing technologies need to show a dramatic reduction in cost compared to the traditional technologies [10].
- In most cases, the low cost of R2R manufacturing can only be exploited if the facility is operating close to the full production capacity. In R2R processing the challenge is in the limited variety of products that can be run, and the large capacity of any one facility. The variety of products is limited because the sequence of process steps is fixed. This is in contrast to a typical semiconductor fabrication, where the individual pieces of automated equipment stand alone and multiple process sequences are supported . The advantage of multiple process sequences is that a much larger number of products can be manufactured in the facility helping to keep it fully utilized. [7]
- The other challenge is that the low cost of R2R results from the rapid process time, that means that even more production is needed to fully utilize the facility. Therefore an application must have very high volumes and /or large areas to utilize a R2R fabrication supporting a single process sequence. Solar cells and display films are two applications where the devices are large area and the potential markets are also very large [7].

- The infrastructure for manufacturing large area flexible displays does not yet exist, so factories wishing to incorporate R2R processing technologies would have to deal with very high start-up costs due to custom-built tools [10] [17].
- As R2R manufacturing processes transcend from fundamental science to laboratory-scale production platforms within academia and industry, the transition to pilot-scale production is hampered by several factors that can benefit from standards. The resulting delay in the commercialization cycle includes losses resulting from incoming materials variations, process technologies, process tolerances, equipment and operator inconsistencies, and lot-to-lot variations, making it extremely costly and difficult to scale to R2R production capacity. Standards are necessary to assist in the translation of discrete processes to an integrated manufacturing flow [1].

2.4. Public and private activities to date

The “additionality” for R2R manufacturing resides in many entities. Government agencies (DOD Army, NSF, DOE Offices), national labs (LBNL, ORNL, NREL, PNNL, etc.) large companies (PlasticLogic, POLYIC, Philips), and academia (University of Mass Amherst, University of Kentucky, Binghamton University) have current interests in energy saving technologies that can be produced by R2R manufacturing processes.

A 2013 report by Information Handling Services (IHS) stated that “of 483 roll-to-roll processing technology patents, 23 flexible OLED-related U.S. published/issued patents and 9 international patents were extracted as key patents. Looking at the application trend of 483 patents (Figure 11) on roll-to-roll processing technology, the number of applications has continuously increased since mid-2000s, and many were applied in the U.S. Major applicants include 3M Innovative Properties, SiPix Imaging, Fuji Film, and General Electric. Amid vigorous developments of roll-to-roll processing technologies, competition among companies in the U.S., Japan, and South Korea gets increasingly fierce.” [18]

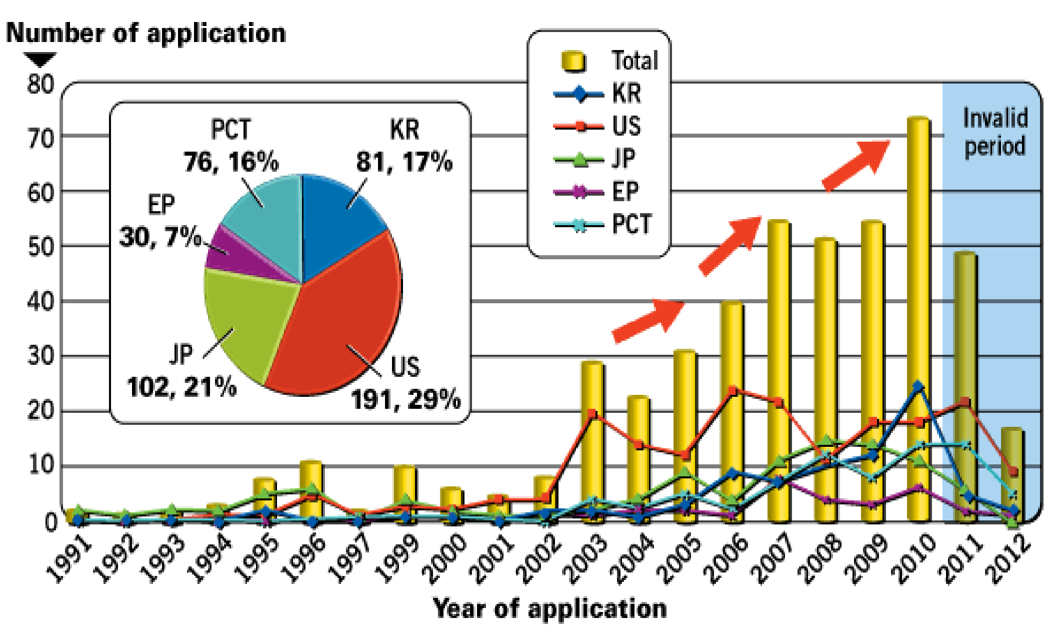

Figure 11 - Roll-to-roll Processing Technology Patent Application Trends by Year/Country [17]

Although R2R processes have been used in various applications for decades, very few patents have been filed until the early 2000's, as can be seen in Figure 11, and is steadily increasing over the last decade.

2.4.1. Current Research Efforts by DoD

The combined services, including the Air Force Research Laboratory (AFRL), t the Army Research Laboratory (ARL) and the Naval Research Laboratory (NRL), have all been actively cooperating in research on micro-electronics, focused on flat panel displays [19].

- More recently, ARL has sponsored research in thin film transistor arrays for displays and digital x-ray detectors. Currently, ARL manages The Flexible Display Center (FDC), based out of Arizona State University. The FDC is a unique public-private partnership with the goal to accelerate the availability of the flexible display technology for the Soldier (FY04-FY13). Some of the results of the FDC work, include demonstration of the world's first flexible electrophoretic display (E-ink Corporation) using the ASU patented bond-debond manufacturing process, development of the ultra-large format flexible full color OLED displays (14.7" diagonal) and x-ray detector arrays (FDC-Defense Threat Reduction Agency (DTRA) and the Palo Alto Research Center (PARC))), a range of hand-held devices with an integrated flexible reflective displays (E-ink Corporation), flexible reflective displays used in an Army field experimentations (Physical Optics Corporation) and the fully flexible tablet for Soldier experimentations (Physical Optics Corporation), flexible microelectromechanical systems (MEMS), among others [20]. Figure 12 illustrates an R2R processed Silicon Radio Frequency Identification (RFID) chips. [21]

Figure 12 - Silicon Radio Frequency Identification (RFID) chip with antenna processed using R2R technology [20].

- Further, starting in FY11, DTRA and ARL are collaborating on developing flexible digital x-ray detectors using the display manufacturing process, although currently the manufacturing process is plate to plate lithography, R2R is under consideration.
- A project with Hewlett-Packard (HP) and PowerFilms which was designed to advance plate to plate and R2R, Self-Aligned Imprint Lithography (SAIL) process for display applications based on amorphous silicon (Si) thin film transistor (TFT) arrays. Although the program was concluded without any commercialization of technology in FY11, process feasibility was demonstrated. Effort is now continuing via an ARL and FlexTech alliance focused on SAIL development.
- The ARL has investments through the FlexTech Alliance, a 30 industrial member consortium of both domestic and international organizations. Their focus includes work on zinc-polymer battery chemistries (referred to as Imprint Energy) that can be processed using screen printing fabrication approaches. Using some of the more mature zinc-polymer chemistries, battery process development has advanced enough to demonstrate prototypes which provide

reasonable performance. The effort also included TFTs and R2R processed OLEDs among others. The FlexTech Alliance also sponsored flexible Si complementary metal–oxide–semiconductor (CMOS) chips on paper, soldier health monitoring systems and other electronics designed to provide and enable prognostics and diagnostics. Many of these exploratory programs are at a Technology Readiness Level (TRL) 1-3. The paper-based flexible Si project is further advanced at a TRL 6 and Manufacturing Readiness Level (MRL) 3-4. This project represents a non-traditional flexible Electronic Manufacturing Services (EMS) program.

2.4.2. Current Research Efforts by DOE

The DOE supports research and development (R&D) in the area of fuel cells, energy efficient buildings, solar energy, batteries and electric vehicles, advanced manufacturing technologies and fossil fuel energy as part of a broad portfolio of activities to secure the nation's energy future.

- **The Fuel Cell Technology Office (FCTO)** develops fuel cells, which use fuels from diverse domestic resources to generate electricity efficiently, and hydrogen, a zero-carbon fuel when produced from renewable resources. These technologies comprise key elements of the DOE portfolio. Fuel cells address energy security by reducing or eliminating oil consumption in transportation energy generation applications. Fuel cell electric vehicles (FCEVs) operating on hydrogen from distributed natural gas can almost completely eliminate petroleum use. At 25% market penetration by 2050, FCEVs can reduce consumption of petroleum by more than 420 thousand barrels per year (Mbbl/yr) compared to 435 Mbbl/yr consumed by the same number of internal combustion engine vehicles (ICEVs). Fuel cells can also provide highly reliable grid support; for example, during Hurricane Sandy, 22 of the 23 400-kilowatt (kW) United Technologies Corporation (UTC) Power (now ClearEdge Power) stationary fuel cells in New England and New York provided continuous power to buildings.

  Assuming 15,000,000 fuel cell vehicles are manufactured per year (10% of the world market in 2030), 4.5 billion membrane electrode assemblies per year produced at a rate of 11,700 membrane electrode assemblies (MEA)/minute are needed for the fuel cell stacks. To achieve a quality requirement for MEAs of 0.1% stack failure, only one critical MEA failure in 300,000 would be allowed; and for six sigma stack quality, only one critical MEA failure in ~90 million would be allowed. Quality control (QC) is critical and tools are needed. However, efforts like these exemplify **an "enduring economic benefit"** for both the public and commercial sectors.

  As an example of R2R manufacturing challenges, Ballard Material Products (now AvCarb) was funded to develop a continuous mixing and coating process to manufacture gas diffusion layers for polymer electrolyte membrane fuel cells. Enhancements to the coating line included modified solutions (e.g. increase solids to reduce wet-load) and optimized dryer profile utilizing dew point sensors to prevent premature drying of the top layer. Using modified slot heights and a multilayer coating head, defect-free coatings and improved cross-web-basis weight uniformity resulted in successfully-produced defect-free anode and cathode materials. Improved repeatability of basis weights was achieved by installing Micro Motion flow meters for each solution. Issues still to address include formation of small agglomerates in the in-line ink due to solution modifications, trade-offs between modified solutions and mix quality, high amount of entrained air present with in-line ink, and examination of methods to improve a de-gas technique to remove air more efficiently, Bottom line is that gas diffusion layer (GDL) costs have been reduced over 50% since the start of the project and Manufacturing capacity has been increased nearly four-fold since the project began.

There is a **"proper role of government"** in transitioning technology to the commercial sector. One example was through the DOE Market Transformation Appropriations and the American Recovery and Reinvestment Act (ARRA). DOE successfully deployed nearly 700 fuel cell material handling units with such customers as FedEx, Sysco, and Whole Foods. These deployments led to almost 5,400 industry funded and "on order" units with no DOE funding. The ARRA investment for these fuel cell powered lift trucks is about $9.7M with an industry cost share of $11.8M. ARRA support was used to demonstrate the commercial competitiveness of fuel cell backup power for telecommunications with over 820 fuel cell units and more than 80 units from Market Transformation Appropriations. As a result of government funding, sales of these technologies continue to grow without federal support with almost 3,600 industry-funded and "on order" fuel cell units for backup power.

- **Solar Energy Technologies Office (SETO)** (through the SunShot Initiative) invested $30 million (with 50% cost share matching) in establishing a consortium called the U.S. Photovoltic Manufacturing Consortium (PVMC) in Albany, New York to support copper-indium-gallium-selenide (CIGS) photovoltaic (PV) products. Initially, a Manufacturing Demonstration Facility (MDF) was established for manufacture of CIGS on a steel web. US-based companies (Global Solar, MiaSole, NuvoSun and Ascent Solar) were interested; however, the dramatic price decrease of conventional crystalline silicon photovoltaics has led to several other U.S.-based R2R CIGS start-ups going out of business over the last two years. There has also been Asian acquisition of all the companies still in business in the United States (except NuvoSun).

  U.S. companies, still working in the CIGS R2R area, are not prepared to use the PVMC MDF and share what they consider to be proprietary processes in the context of a consortium. This is an example where "openness" was not a contributing factor. Public announcements indicate that these companies intend to scale in Asia and other developing nations. As a result, SETO has redirected the consortium to work on "downstream" issues in support of flexible CIGS, such as establishing the methods for installation of flexible PV modules and determining the reliability of flexible PV.

- **The Building Technologies Office (BTO)** has a number of existing Investments in R2R manufacturing including architectural applications research with Lawrence Berkley National Laboratory (LBNL) regarding airflow panel membranes, with Oak Ridge National Laboratory (ORNL) in R2R sensors for building applications, with the National Renewable Energy Laboratory (NREL) investigating VI window film, with ITN Energy Systems and the Electric Power research Institute (EPRI) work on Low-energy/Electrochromic window film, with 3M/LBNL investigating daylighting film for windows, with 3M/ORNL (within the China Clean Energy Research Center (CERC) program) work focused on primer-less, self-adhered air sealing membranes, with PPG/Pacific Northwest National Laboratory (PNNL) developing infrared (IR) responsive window coatings, and with Heliotrope Technologies work on near IR Electrochromic (NIR EC) window coatings.

  The Advanced Manufacturing Office (AMO) (through prior programs supporting Inventions and Innovation as well as Industrial Sensors and Small Business Innovation Research (SBIR)) invested approximately $1 million in cadmium-tellurium (CdTe) solar cell development and manufacturing; approximately $1 million in advanced solar-reactive glazing, coating and manufacturing technologies to reduce unwanted solar gain through windows, skylights and automotive windows; approximately $1 million among battery technologies, super-capacitor technologies, superconducting cable technologies; and approximately $2M in advanced sensor technologies. AMO investments focused on lithium-ion (Li-Ion) battery technology incorporating R2R processing in the effort. A MDF has been established at ORNL with focus on electrolyte materials used in laminated planar battery pack assemblies.

- **The Office of Fossil Energy (FE)** has investments concerning CO_2 membranes. Those involving R2R manufacturing processes are being considered or used to manufacture several different polymeric and ceramic/metallic membranes for CO_2 separation for power plants. Similar processes are used to manufacture existing commercial water filtration and natural gas processing membranes. FE has not been investing in the commercial production of membranes, but rather left the commercialization of the materials to the project performers and their partners. Issues such as defect control during coating and drying, substrate and active layer bonding, and quality control/quality assurance (QC/QA) are consistent issues with manufacturing CO_2 membranes. Many of the technologies are at the pilot scale and much of the manufacturing processes efforts are considered at a similar scale of development (TRL 4-5). The investments in membranes detailed in Table 2 for post-market and pre-market applications have been made to date and may benefit from a concerted effort to improve the R2R manufacturing processes.

Table 2 - Office of Fossil Energy Investments in CO_2 Membranes

Application	Company/Agency	Substrate	Active Layer	Type
Post	Ohio State University	Polymer - Polyethersulfone	Zeolites	Spiral Wound
Post	Membrane Technology & Research, Inc	Polymer	Polymer	Spiral Wound
Post	General Electric	Polymer	Phosphazene	Hollow Fiber
Post	Gas Technology Institute	Poly ether ether ketone (PEEK)	Perfluoro-oligimer	Hollow Fiber - Gas/Liquid
Post	Argonne National Laboratory	Alumina-Zirconia	Pd/TZ-3Y cermet	Long-tubes
Post	Pacific Northwest National Laboratory	Ceramic/Metallic	Ionic Liquid	Sheet/Plate
Pre	Praxair	Ceramic	Pd alloy	Shell and Tube
Pre	Eltron	Metal Alloy	Not Applicable	Shell and Tube
Pre	Worcester Polytechnic Institute	PSS-316L	Pd Alloy	Shell and Tube
Pre	Pall Corporation	Ziconia Coated SS Tubes	Pd Alloy	Shell and Tube
Pre	Los Alamos National Laboratory	Polybenzimidazole (PBI) - Polymer	PBI Polymer	Hollow Fiber

- **National Renewable Energy Laboratory (NREL)**
  NREL efforts developed a defect diagnostic in house by applying a direct current (DC) potential to a membrane electrode assembly and then monitoring the heat generated in the MEA using

an IR (heat) detector. Areas in which there is a defect generate no heat and no signal for the detector. NREL demonstrated the IR/DC technique on Ion Power's production coating line for detection of electrode (on decal) defects.

R&D at NREL is addressing quality control needs for scale-up of fuel cells and cell component manufacturing on weblines. The approach includes understanding quality control needs from industry partners and forums, developing diagnostics, using modeling to guide development, using in situ testing to understand the effects of defects, validating diagnostics in-line, and transferring technology to industry.

- **National Science Foundation (NSF)**

  NSF supports fundamental and translational research efforts within the Center for Hierarchical Manufacturing, an NSF-supported Nanoscale Science and Engineering Center (NSEC), leveraging $4 million/year of federally-funded nanomanufacturing research, The research program focuses on the integration of nanofabrication processes for 30 nanometer and smaller elements based on directed self-assembly, additive-driven assembly, nanoimprint lithography, high fidelity 3-D polymer template replication, and conformal deposition at the nanoscale with Si wafer technologies or high-rate R2R-based production tools.

  NSF also supports fundamental and translational research efforts within the Nanomanufacturing Systems for Mobile Computing and Mobile Energy Technologies (NASCENT), an NSF-supported Engineering Research Center (ERC) leveraging $4 million/year of federally-funded research on innovative nanomanufacturing, nanosculpting and nanometrology systems that could lead to versatile methods for the high-volume nanomanufacturing of mobile computing devices such as wearable sensors, foldable laptops and flexible batteries.

- **Other**

  Over the last 10 years, the European Union has had significant investments in R2R manufacturing and related printing plate-to-plate using organic based TFTs for displays and RFIDs. Some organizations involved, include: PlasticLogic (focused on plate-to-plate), POLYIC (involved in R2R RFID), and Philips and one of its subsidiaries, PolymerVision. The former company has recently introduced a range of flexible electronic OLED displays designed as "wearable" devices.

  Commercial alkaline battery manufactures focus on processes using R2R process techniques. Goal is to "build" structure, which includes "can" material, anodes and cathodes in a continuous process, with individual assembly achieved via a mechanical formatting operation at the end of the R2R process.

  The University of Massachusetts (UMass Amherst) within the Center for Hierarchical Manufacturing sponsors the "Research Cluster R: Roll to Roll Process Research Facility. The facility supports efforts focused on nano-imprint lithography (NIL) process and development. Current investment allows work up to 6-0inch wide format using a range of R2R equipment and analytical tools. Focus areas include; planarization, imprint embossing and patterning, alternative materials and membranes, functional hybrids, viscoelastic fluids, R2R integration and design for manufacturability.

The University of Kentucky Center for Applied Energy Research has a significant effort underway which focuses on a range of energy applications, some of which involve R2R. Areas of interest include low-cost carbon anode precursors, VRF, Thermoelectrics, etc.

Flexible "heater" circuitry for displays has been commercially available from companies such as All Flex Flexible Circuits, LLC for over 25 years. These products are fabricated using a mixture of R2R and batch "plate to plate" techniques, involving micro-electronics lithography printing and chemical etching processes.

The Center for Advanced Microelectronics Manufacturing (CAMM), a partnership between Binghamton University (BU), Endicott Interconnect Technologies (EI), Cornell University and the Flex Tech Alliance, is a prototype R&D facility in large area flexible electronics. The CAMM is part of BU's New York State Center of Excellence in Small Scale Systems Integration and Packaging (S3IP), which serves as an international resource for systems integration and packaging R&D.

2.5. R&D in R2R Processing

2.5.1. Technological Needs of R2R Processing

- **Providing a connection between emerging R2R process R&D and scaled manufacturing**: In order to extend R2R manufacturing technologies to volume manufacturing, several infrastructural needs must be established relevant to emerging processes and tools [1].

    - R2R manufacturing traditionally consists of coating and printing processes. While there are many companies engaged in R2R manufacturing, there remains a general lack of standardized infrastructure in some cases, and most academic institutions do not have R2R fabrication facilities. As a result, R&D data are still lacking on what are achievable with R2R processes and what are the limitations, especially in the context of throughput [1].

    - Parameters affecting throughput and defects control for various processes need to be established. In addition, the necessary supply chain is lacking, and needs to be broadly established, along with standards. Standards developments cannot be underestimated, and a more concerted effort in this area is necessary. One way to address these issues may be through the establishment of pilot line facilities for development, demonstration and optimization of full processes. This infrastructure component would provide a vital step between lab coupon-scale development and production line scale-up, ultimately reducing risk and cost, and providing a more rapid development path for product commercialization. This is due to the high cost of roll-to-roll instruments. This can be alleviated by creating shared facilities (such as the Research Cluster R for Roll-to-Roll Processing at UMass Amherst) that will be accessible for academics and for industrial participants to try out some ideas, as well as establish emerging processes and materials for broader use [1].

    - Previous DOE sponsored workshops [22] have identified a need to address equipment and quality issues. Equipment needs to support formats sufficient to meet nano-scale atomic layer deposition (ALD) and small-scale (e.g. microelectronics thin and thick-film)

to medium-scale (windows and window films) to large-scale (membranes for biofuel and natural gas processing) fabrication.
Investigations for manufacturing development should focus on tools to feed pre-requisite solutions, slurries at sufficient rates while controlling rheologies of these materials, webs (tensile strengths, surface finish and release, materials, zero defect, etc.), motor controls, motors (web speed control, tensioning, material "take-up", post formatting, etc.), metrological instrumentation, simulation and design tools, control(s) feedback and adjust, materials drying accessories, ventilation and effluent treatments, incorporation of concurrent/simultaneous process using additive and subtractive, in-air, other atmosphere and vacuum processing, precision alignment, lithographic imaging and etch/deposition, etc.

- **Tackling challenges related to process tools and core capabilities:** [1] These include:
    - Large-area, cost-effective e-beam patterning tools/capabilities
    - Plasma etching tools for large-area, uniform R2R processing
    - Ink jet applicators compatible with wide range of UV monomers
    - Development of high-quality nickel metal electroforming processes for high aspect ratio, large pattern volume structures
    - High-durability, low-cost transparent imprinting of molds, or, inexpensive/fast replacement transparent molds
    - Fabrication of seamless cylindrical imprint molds
    - Large-area, real-time metrology and process characterization

- **Further developing emerging process tools towards large area processes**: [1]
    - Precision ink jet fluid applicators
        - Any dot, anywhere; high uniformity and thickness control
    - Atmospheric plasma etching
        - Lower-cost surface processing with elimination of vacuum step
    - R2R-ALD
        - Precision application of very thin layers at high rates
        - High conformality; uniform coating of aspect ratios up to 1000:1
        - High film density, low film stress
        - Continuous, pinhole-free ultra-thin films

- **Developing imprint and web materials**: [1] Materials are critical for the extension of R2R manufacturing processes to large area and high throughputs.
  In nanoimprint technology, the imprint materials need to be developed (especially for UV roll-to-roll imprint). There is also a limited supply of suitable web materials. Examples for a "materials wishlist" include:
    - UV polymers that resist plasma crosslinking
    - Transparent conductive polymers having:
        - Higher conductivity and light transmission
        - Improved durability/stability
        - UV curable
    - Less costly, higher-temp substrates
        - (>250C; preferably clear…)

- **Tackling metrology and instrumentation challenges**:

    - Commercial enterprises which incorporate R2R into their product process routes have serious control issues regarding means to detect, control, and otherwise eliminate potential quality issues within products prior to investing additional value add. Cross-cutting needs include items such as; thickness measurement, inspection for mechanical defects such as pinholes and cracks, measurement of electrical properties such as resistance measurement of surface texture, structure and morphology, inspection for inter-layer delamination and voids, etc. Programs are highly desirable that generically investigate these issues with respect to differences in scale, criticality, application, ex-situ measurement while advancing tools and methods for the collection, analysis, storage, and use (either in real time or for later data mining) of high volumes of in-line QC data, and for the integration of these data into process control and feedback systems. Ultimately determinations of means to predict/correlate defects to performance would be one of the prime measureable program metrics.

    - Defects are undesirable for printed electronics since they cause open and short circuits, thus destroying the performance of devices. There are several factors that cause defects such as missing nozzles in the print head, particles on the substrate, particles on the screen/stamp, web wander, non-uniform web tension, mis-registration, etc. A few examples of defects are shown in Figure 13.

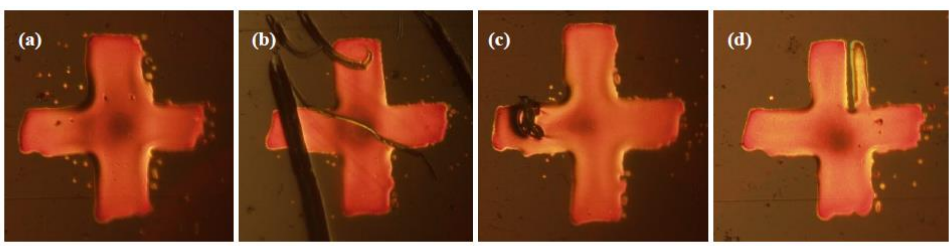

Figure 13 - Optical microscope images of (a) an intended pattern, and (b-d) show defects in the pattern [23]

    - Metrology and inspection incorporating in-line optical techniques are presently being developed, but significant challenges remain for monitoring of high throughput processes having nanoscale features. In combination with this, model-based real-time diagnostics and control would complement the development of process modeling and control methods [1].

    - For high rate R2R manufacturing, inspection and quality control is a critical area that determines successful outcome. These include defect detection, surface roughness measurement, inspection of layer quality, measurement of electrical properties to ensure proper functionality, registration control, possibility for repair/correction, product testing, etc. [23].

- Metrology and instrumentation challenges include availability of particulate-free high quality substrate, development and implementation of high-speed in-line and off-line inspection and diagnostic tools with adaptive control for patterned and unpatterned material films, development of reliable hardware, etc. These challenges need to be addressed and overcome in order to realize a successful manufacturing process. Due to extreme resolution requirements compared to print media, the burden of software and hardware tools on the throughput also needs to be carefully determined. Moreover, the effect of web wanders and variations in web speed need to accurately be determined in the design of the system hardware and software. [23]

- Realization of successful metrology and instrumentation by overcoming the challenges for the development of a R2R manufacturing system for flexible electronic systems opens limitless possibilities for the deployment of high performance flexible electronic components in a variety of applications including communication, sensing, medicine, agriculture, energy, lighting etc. [23].

- Metrology, standards and inspection requirements for R2R are [24]:
  - defect inspection-pattern defects
  - characterization/pattern inspection
  - laser scattering/particle size distribution
  - final yield as means to identify defects
  - cost involved for now at micron scale for adapting tools to R2R web platforms
  - smallest features inspected can reach to 1 µm.
- A summary of industry inputs on in-line QC techniques directly from EERE's Quality Control Workshop, which was held in 2013 in Golden, Colorado [22] are as follows:
  - Techniques currently used in industry to identify and quantify defects in materials are:
    - Vision detection systems for cracks
    - Fluorescence of functional coatings applied to textiles
    - Non-contact eddy current measurements for surface sheet resistance
    - Non-contact optical measurements for band gap and relative thickness of coatings
    - Non-contact x-ray fluorescence (XRF) for composition and also thickness of coatings
    - Photo-imaging for physical defects
  - Existing issues with the current quality assurance/quality control techniques
    - Lack of standards. A few companies sell cameras and algorithms, but not necessarily tuned to the application
    - Hardware exists. Main gap is software relevant to specific application
    - Need to be able to scan for the composition of coatings (for multi-material coatings) and physical defects across full width and length of web while web is in motion
  - Measurements needed for in-line quality control that current techniques do not address

- Physical defect density and/or pinhole density
- Band gap measurements
- Surface sheet resistance of coatings
- Optical transmission
- Relative thickness of coatings across and along the length of the web
- Material composition measurements
- Networking-cloud data transmission

2.6. Emerging Processes and Tools for R2R

2.6.1. Atomic Layer Deposition (ALD)

ALD is a thin film deposition technique in which films are grown by the sequential pulsing of chemical precursors onto the surface of a substrate. A typical process sequence involves introduction of precursor A, followed by a system purge, then introduction of precursor B, followed by another system purge, after which the steps are repeated. The precursor reactions on the substrate surface lead to the growth of the thin film on a layer-by-layer basis, with the resulting film thickness controlled by the number of cycles of the process sequence. As the deposition process is self-limiting, the films are extremely uniform, pinhole free, and exceptionally conformal. ALD is capable of depositing a range of metal oxide films, as well as a limited number of metal coatings, and has the further advantage of relatively low-temperature processes and reasonably low-cost precursors for most applications. As a result, ALD is finding traction in the semiconductor industry, and has further been scaled to large-area substrate processes for thin film photovoltaics and displays where the metal oxide coatings yield superior barrier coatings and dielectric films [1].

2.6.2. Potentiometric Stripping Analysis for Electroplated Alloys

Electroplating represents an additive, solution-based deposition process suitable for R2R platforms for a range of metals and alloys. A key challenge for continuous, high-speed coating systems is the control of stoichiometry and the depletion the plating baths in web-based systems. The potentiometric stripping analysis (PSA) techniques precisely control both stoichiometry and uniformity of metal and alloy coatings on flexible webs. Keys to maintaining sufficient process control in the electroplating steps included keeping the solution at the work surface fresh and evenly biased by agitating the bath, providing adequate circulation, further utilizing an inert environment such as an argon blanket to minimize the effects of oxidation, and utilizing a separate anode for precise control of field distribution [1].

2.6.3. High Temperature R2R Processes

ORNL is conducting the research in the development of high-temperature R2R processes suitable to create crystalline, high-performance semiconducting materials. The high-temperature process capability can exceed 1200°C through the use of a high-temperature metal or suitable substrate; calendaring of thin film coatings then occurs, followed by a thermal pressing step. Because typical processes tailor a series of functional layers, inter-diffusion becomes a significant concern. This issue is resolved by depositing a stack of buffer layers to provide the required crystal orientation that include deposition of a diffusion barrier and then active layer coating. The high-temperature R2R processes can be used to develop hybrid solution-based approaches as well. The high-temperature processes are suitable for a range of thin film crystalline materials, including silicon for solar PV, diamond, and other semiconductors. This R2R process capability opens up opportunities for large-area, high-quality semiconductors having electronic transport properties approaching those of bulk materials, thereby enabling high-performance electronic devices and systems [1].

2.6.4. Standards Development

Standards are an important aspect of the successful commercialization of any product, and typically are underestimated. Benefits of standards include building end user confidence, creation of a common language between producers and users, promotion of product compatibility and interoperability, overcoming trade barriers to open markets, and fostering diffusion and adoption of technology. As printed electronics and R2R manufacturing transcend from fundamental science to laboratory-scale production platforms within academia and industry, the transition to pilot-scale production is hampered by several factors that can benefit from standards. The resulting delay in the commercialization cycle includes losses resulting from incoming materials variations, process technologies, process tolerances, equipment and operator inconsistencies, and lot-to-lot variations, making it extremely costly and difficult to scale to R2R production capacity [1].

2.7. Key Technology/Application Opportunity Areas

2.7.1. Membranes

Areas of interest might include (but not limited to); high pressure "ceramic" membranes, indoor air quality and dehumidification membranes for applications in buildings, other water processing, gas separations for natural gas processing and CO_2 capture application s (CO_2/N_2, CO_2, H_2, and CO_2/CH_4), and liquid/gas separation membranes (CO_2 loaded solvents), forward osmosis capacitive polarization membranes, and other multilayer systems such as those used in battery applications, i.e. VRF which support high permeability rates, resist reject material "buildup" and are environmentally "friendly". Current production cost of membranes is ~$100/m^2. Adapted manufacturing processes used by the RO industry are needed to reduce costs by at least 50%. Current membrane market is $16.5B globally – United States demand is approximately $1.7B total - Liquid separation is $1.5B and ~$0.15B for gas separation. Expected to rise ~7% per year. Global membrane demand is expected to be $25.7B in 2017 and continue to rise ~10% per year [25].

2.7.2. Advanced Deposition Processes

Formatted, higher quality depositions are needed. Equipment needs to support formats sufficient to meet microelectronics to building sector requirements. Investigations/development will focus on tools to feed solutions and slurries at high rates while controlling solution rheologies; web properties , motor controlsfor web speed control, tensioning, material "take-up", post formatting; metrological instrumentation; control feedback and process adjustment; materials drying; ventilation and effluent treatments, and incorporation of concurrent/simultaneous processes. Emphatically, all of the aforementioned is needed at all-size scales, i.e. from the nano/atomic scale through thin-film to thick-film size-scale.

2.7.3. Flexible Electronics

An intrigue space is R2R additive manufacturing for EMS. The technology applications are well defined, most of the materials are well defined in manufacturing, mostly the technologies, with exception of interconnects that are mature (MRL 4-5). However, government investments are necessary to reduce risk for industry to participate. EMS is a $300B/year industry. The market is in printed circuit board population with Si CMOS and passive components. On a limited basis, R2R is used in single chip integration for smart labels (RFID tags and antennas), such as products offered by Muhlbauer High Tech International [26]. Another area of possible manufacturing development involves OLEDS which can be processed on flexible substrates [27], as shown in Figure 14. Systems have been developed which have exhibited a brightness as high as 10,000 candela per square meter. DOE projects the benefits of replacing traditional systems with phosphorescent OLED lighting, in the time frame of 2012 to 2018, as reducing energy use by 0.22 quadrillion Btu's, saving domestic consumers $20 billion and reducing environmental pollution emissions by 3.7 million metric tons [28].

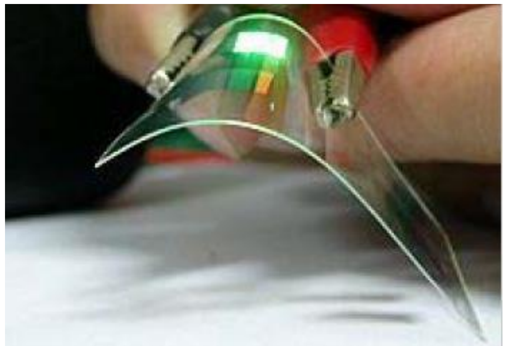

Figure 14 - Demonstration of a flexible OLED device. Photo: General Electric

The second area of interest focuses on larger format flexible displays, detectors and other sensors, such as used for neutron and other E-M arrays. This path will be to move from plate-to-plate standard lithography as used in the industry to continuous R2R processing. This approach leverages $90M of existing U.S. Army investments, and $1T+ of industrial private sector investments in traditional flat-panel glass manufacturing. The current TRL levels are 3-4 for emerging applications, TRL 5-6 for the maturing flexible digital x-ray technology. The MRL level is 5, with development necessary to broaden the application space, reduce cost and improve yield. Here the work would attempt to merge traditional processes with some R2R technology. If a major thrust involves sensors, opportunity to continue development of materials (incorporating new material sets, enhanced efficiency of traditional types, i.e. substitutional elements, enhanced process, etc.) will be investigated. Sensor efforts include from MRL/TRL 1 to fully commercialize. Those efforts within the MRL/TRL 4 to 7 include those that would serve to incorporate program information to feed MetaData collection, design of more efficient/selective devices, develop imbedding processes within other materials-assemblies, means to enhance signal processing, and collected data "Cyber Security". This latter being needed as users will need to collect data remotely to gage quality, state-of-the condition to enable state of process or mean-time to failure, response characterization, etc. via the "internet cloud" to be successful. If one considers thin-film MEMS sensors and devices within the scope of this sector, the market could exceed $1 trillion.

A third area would be to focus on the advancement of materials with associated equipment to enable commercialization of NIL and patterning with 50 to 100nm print resolution at process rates of 3 to 5 meters per minute. Fourthly, flexible electronics need to include investigation of lighting technologies. This would include moisture and environmental barrier materials/layers with ALD of LED and OLED technologies with associated packaging systems, which are fabricated in multilayer fashion to achieve a hermetic, moisture-proof package.

The current research work is focused on the following topics: [29]

- Developing roll-to-roll manufacturing of thin film electronics on low-cost flexible substrates using Pulse Thermal Processing (PTP) technologies coupled with non-vacuum low temperature deposition techniques.
- Developing non-vacuum, large scale deposition and processing techniques for nanoparticle-based inks and pastes that reduce cost and energy requirements associated with processing of thin film electronics.

- Ink development and annealing studies to increase the crystallinity and photo luminescent efficiency of Zinc-Gallate coatings.

2.7.4. Battery Technology

Including existing agency investments and commercial development results, work should focus on a wide range of battery chemistries, i.e. Li-ion, Zinc-polymer, Li/CFx, Vanadium Redox Flow (VRF) systems, and advanced alkaline systems. Continuous materials deposition on webs to build the "multi-layer" configuration using tape cast, screen print, vapor or wet chemical deposition or evaporative/sputter techniques could be included. Of special interest would be deposition of carbon nano-tubes and whiskers on graphene for certain applications.

The current research work to apply R2R processes in flexible thin-film battery manufacturing is focused on the following topics: [30]

- Reducing excessive scrap rates of electrode coatings.
- In-line quality measurement and control – For example: In-line laser sensing for thickness monitoring, in-situ materials diagnostics with ex-situ structural characterization.
- Reducing manufacturing as well as associated system cost by implementing in-line Non-Destructive Examination (NDE) and QC].
- Scaling-up, Industrial issues of yield and throughput.

2.7.5. PEM Fuel Cells

The manufacture of fuel cell stack components utilizing continuous, high volume, lower cost process technologies is needed. Current R2R technology and methods need to replace the manual preparation of layers, such as painting catalyst ink by hand onto decals that are then thermally pressed onto membranes. The process should also address high-speed sealing of assemblies which can be accomplished using R2R processes. Figure 15 shows an approach that WL Gore & Associates [31] is working on to coat electrodes directly onto membrane material saving steps and material thus saving money.

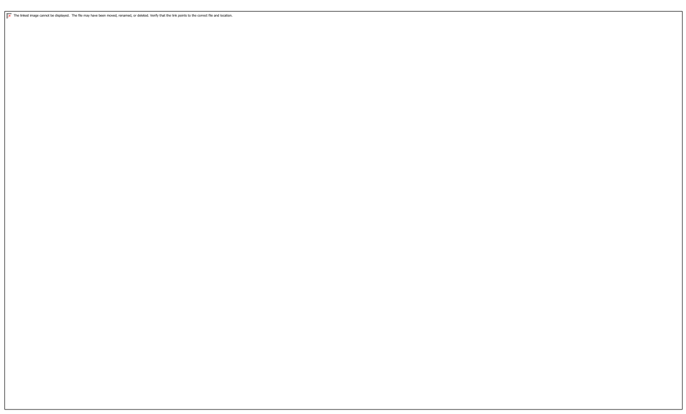

Figure 15 - Approach to coat electrodes directly onto membrane material saving steps and material

In 2012, the domestic fuel cell and hydrogen energy industry was expected to produce $785 million in revenue [32]. Funding from DOE EERE for hydrogen and fuel cell R&D has played a critical role to enable this emerging fuel cell industry. According to Bloomberg New Energy Finance [33], DOE funding was approximately equal to venture capital and private equity investment in the United States in 2011. EERE funding has led to more than 450 patents, 40 commercial technologies, and 65 emerging technologies for hydrogen production and delivery, hydrogen storage, and fuel cells.

2.7.6. Photovoltaics

- In addition to efficient processing, efficient process control during manufacture is required, and new materials and processes are urgently needed. Some of the most important materials and processes are those that will enable the printing of semitransparent electrodes and complete processes that are built around enabling complete fabrication of efficient solar cells. The materials and processes should of course give access to organic photovoltaic (OPVs) that provide operational stability of more than 10 years and they should be efficient (> 10 %). A particular requirement to the OPV is that it has as thin an outline as possible with low materials consumption, to achieve a low embodied energy. The processing should not be environmentally harmful and should, through use of the lowest possible temperatures, require a very low input energy for manufacture. This will enable short energy payback times. Manufacture of the entire solar cell stack at an overall speed of > 10 m/min will enable the manufacture of a daily energy production capacity of more than 1 gigawatt peak and thus, in principle, fully address mankind's future energy needs [34]. Figure 16 illustrates the assembly of a scalable, encapsulate, large area, flexible, organic solar cell produced by a R2R process. [35]

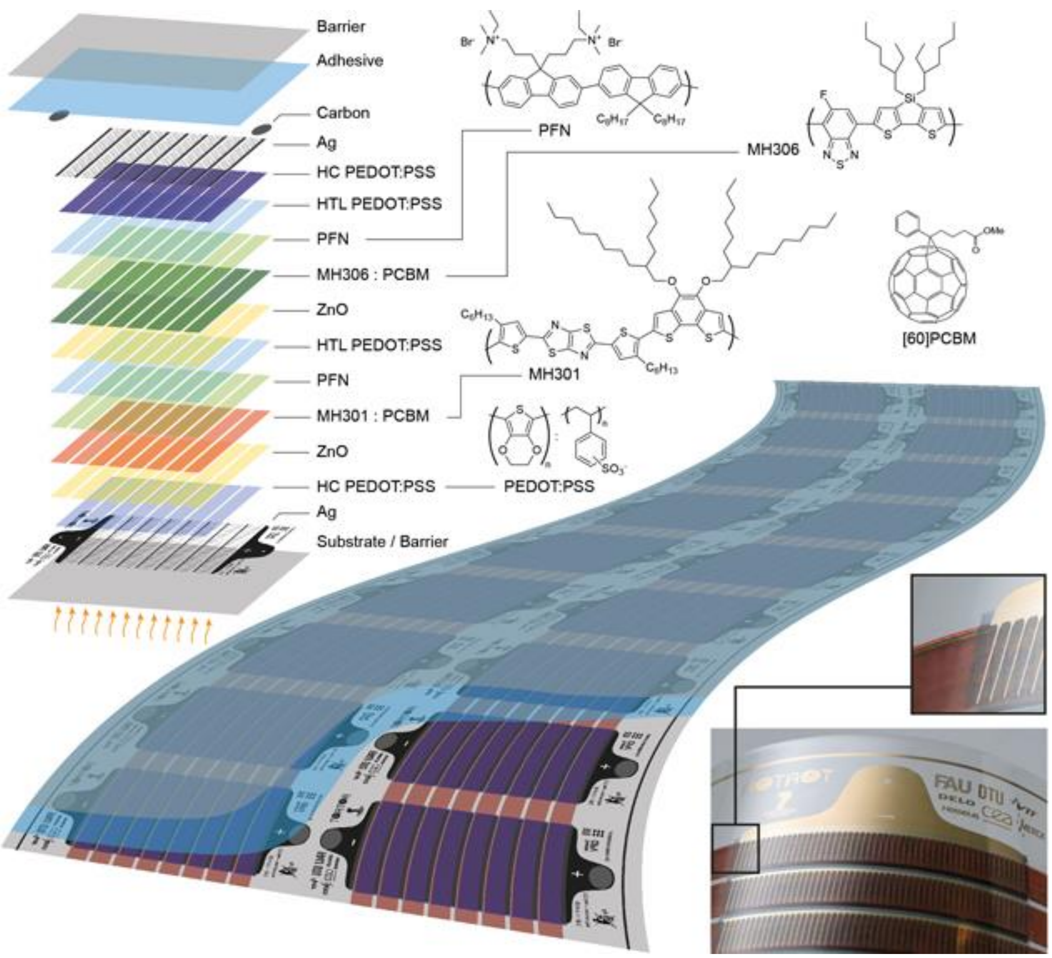

Figure 16 - Scalable, ambient atmosphere roll-to-roll manufacture of encapsulated large area, flexible organic tandem solar cell modules [35].

- The scientific thrust should be with the final form and processing methods in mind and not as it has been until now with a blind focus on high performance in an often unrealistic and not scalable setting [35].

- The active materials and inks needs to be developed specifically with the thermo-mechanical properties of the multi-layer structure in mind. The complex multi-layer structure with different thermal expansion coefficients and moduli for the individual layers and different adhesion energies at each interface are likely to present an enormous challenge for the manufacture of a robust flexible tandem organic solar cell [35].

- Research efforts are needed on the control of film thickness and especially the evenness of the dry films for the multi-layer stack through proper ink design. This involves control of viscosity, ink stability over time, ink rheology during deposition, ink rheology during drying (i.e. heating and up-concentration of solutes in the wet film), wetting behavior during deposition and drying, control over morphology formation and of course it must all work in air [35].

2.7.7. Metrology and Quality Systems

- Commercial enterprises which incorporate R2R manufacturing into their processes must detect, control, and otherwise eliminate potential quality issues within products.

- Technology development needs include inspection for mechanical defects such as pinholes and cracks, measurement of electrical properties such as resistance measurement, and inspection for inter-layer delamination and voids.

- All data would be integrated into process control and feedback systems. These technologies will be used to correlate defects to performance.

2.4.1.1. Embedded Thermal Energy

- There is a need to develop R2R additive manufacturing for electronics applications such as larger format flexible displays, detectors, and stretchable/conformable sensors.

- This technology will lead to a fundamental change for manufacturing these systems from plate-to-plate standard lithography to continuous R2R processing.

2.8. Technology Roadmaps Applicable to R2R Manufacturing

R2R is a type of process, not a technology, and therefore no specific technology roadmap exists for developing R2R processes in general; instead, roadmaps exist for specific technologies that would use a R2R process as the manufacturing method. R2R processes can be improved by insertion of technologies that make the process more efficient and less costly. The International Electronics Manufacturing Initiative (iNEMI) developed a technology roadmap for flexible electronics that addresses materials (nanoparticle suspensions, particle blends, and small molecular solutions), printing technologies (contact and non-contact), and processes (roll to roll, roll to sheet, and sheet). [36] In the United Kingdom, the Centre for Process Innovation developed their technology roadmap to expand R2R and encapsulation processing technologies to target the development of flexible optoelectronic devices for the emerging printed electronics markets and to address many of the challenges encountered in scaling up emerging technologies to commercialization by adopting R2R processing techniques.[37]

From an industry perspective, Baker ™ Wet Process Equipment has developed a technology roadmap to understand and manage the issues associated with conventional versus R2R processing for manufacturing flexible printed circuits. [38] Their roadmap focuses on the core of current manufacturing trends toward producing thinner, lighter and higher density printed circuits by use of effective handling and processing of a thin core material. R2R processing equipment will need to focus on the smooth, yet firm, transport of films through various wet processes in both a horizontal and vertical plane and will require "next generation" spray- or immersion-type technologies. Flexible printed circuit fabrication using batch processing and antiquated rigid-panel processes is responsible for the failure to produce the necessary technologies in the last century and will be superseded by R2R technology in the future. [39] Additionally, the National Aeronautics and Space Administration (NASA) has drafted integrated technology roadmaps for 14 Space Technology Areas, which includes "pull" and "push" technology strategies and considers a wide range of pathways to advance their current capabilities in space. Technology Area 12 addresses materials, structures, mechanical systems, and manufacturing. Although R2R is not specifically addressed as part of the roadmap, several of the technologies and processes, such as hybrid laminates, polymer matrix composites, multi-functional thin films, flexible materials for entry-descent-landing, photovoltaics, lightweight aluminized thin film systems for solar sails and large ultra-light precision optical materials are all directly applicable to R2R manufacturing.[40]

2.9. Workshops on R2R Processes and Manufacturing

Workshops are not held specifically on just R2R manufacturing. Usually a workshop is convened on a technology area such as Nanofabrication Technologies for R2R Processing [second ref] or the DOD and DOE Manufacturing Innovation Topics Workshop [third ref] where R2R manufacturing is an agenda topic or a separate breakout session. Discussions typically focus on using a R2R process for coating of polymer films, device level patterning, imprint lithography methodology, patterning limitations, and NIL for R2R processing of nanotechnologies.[1] Workshops also address programmatic issues for R2R manufacturing such as R2R process technology needs; manufacturing challenges, and investments; process deficiencies and metrological needs; and quality systems and synergy. [41] Workshops are held annually on nanomanufacturing that provide opportunities to share information on emerging processes and scaled manufacturing platforms where R2R may have a role. [42] [43] They can also focus on specific technology areas that have immediate applications to clean energy initiatives such as biomass indirect liquefaction that focuses on pathways that convert biomass-based synthetic gases to liquid intermediates. [44] Inevitability, the common areas of interest lie in overall technology needs, manufacturing challenges, and investment levels.

3. Risk and Uncertainty, and other Considerations

3.1. Risks and Uncertainties of Using R2R Processes and Manufacturing

The risks with using R2R processes are defined in the challenges. R2R manufacturing is not ideal for every type of material manufacturer, but it is ideal for thin and thick film materials with large areas that are required for high volume production with minimal defects and waste. R2R processes, in general, are energy efficient and environmentally friendly. However, as with any type of manufacturing, there are associated risks and uncertainties.

- **High Startup Costs** - A combination of high costs and poor availability of production tools are hindering the adoption of roll-to-roll manufacturing. For example, setting an active-matrix flexible organic light-emitting diode (OLED) substrate line amounted to roughly $177 per square foot. The cost of tooling a passive-matrix polymer light-emitting diode (PLED) line is far less, at $45 per square foot. Still, the long-term promise of roll-to-roll manufacturing is propelling it to the forefront of flexible substrate R&D activity. The Center for Advanced Manufacturing (CAMM), Binghamton, NY expanding its tooling capability to actively research R2R manufacturing for emerging technologies such as large-area LED lighting, photovoltaic cells on plastic, low-cost RFID tags, and lightweight electronics and packaging platforms on rugged, flexible substrates. Also, scientists at Hewlett-Packard Laboratories and Iowa, Thin Film Technologies, are developing large-area arrays of thin-film transistors on polymer substrates using R2R techniques. The approach combines plasma deposition and etching with self-aligned imprint lithography to produce a cost effective end product. [45] Further research in specific applications that employ R2R processes will provide the data needed to reduce the costs of startup.

- **Speed of High Volume/Large Area Process vs Low Volume/Small Item Stand-Alone Process** - The speed and capacity for R2R manufacturing versus a batch process is dependent on the material requirements for the end product and is directly related to costs. As an example, if an assumption is made that conventional operations such as lithography, etching and sputter deposition are used in the R2R process, and 1000-feet by two-feet rolls of polymeric substrate are used to make a final product of 3.25-inch by 3.25-inch LED display on an 18-inch by 24-inch format, then the cost per square foot of active and passive matrix displays are expected to decline with increases of volume. Indeed, studies have shown that the minimum efficient scale for the operation of a R2R display manufacturing facility is around 20,000 square feet per week. Many markets and application areas could support a plant operating at this capacity if the displays could be sold into these markets at a sufficiently high volume to sustain the manufacturing operation. If the plant operated at a capacity of 100,000 square feet per week over a two-year period, the cost of producing LED displays would be about half the cost of a display produced by a conventional stand-alone batch manufacturing approach. Nearly every model in the display industry predicts that a R2R manufacturing facility could offer significant cost savings if it can be integrated successfully. [46]

- **Material Variations/Tolerances/Lot Variations/Scrap** - Variations in substrates and production lots of the end products from R2R manufacturing can be caused by several factors depending on the materials being used, the machinery involved, the control of the web, and the process(es) employed (lithography, deposition, etc.) just to name a few. Even the configuration of the rollers (double side mounted or cantilevered) will produce variations. In some applications such as thin films and nano-materials, the tolerances must be closely controlled in order to get a quality end product. If tolerances and variations are significant, then the R2R process can result in a lot of scrap and waste material that may not be recyclable. This also adds to the cost of manufacture. As the R2R process becomes more adapted to the anomalies in the initial production phases, the material and lot variations are usually reduced and the end products are well within tolerances. Research is needed on various types of instruments can be incorporated into the R2R process to further reduce variations and eliminate scrap.

- **Metrology** - As previously discussed in this section, the success of employing a R2R process in manufacturing a specific technology is heavily dependent on process and cost control. At high rates of R2R manufacturing, metrology is needed to address defects (from static buildup and missing or disconnected patterns), quality of substrate, registration (pattern position), and in-line and off-line optical inspection for quality control. [47] This can go beyond just looking for material defects such as pinholes, non-uniform thickness, and impurities. As an example, at the end of processing polymer solar cells using roll-to-roll methods, one ends up with a roll of material. While some testing can be carried out during the processing of the individual layers, the functionality of the solar cell itself, i.e., the production of electrical energy upon being subject to illumination, has to be carried out at the very end, on the very roll that is the end product. Inline monitoring techniques are useful for guiding the process, but they cannot guarantee the final performance. Therefore R2R instrumentation is also needed to test functionality. The techniques that have proven useful for process control are the camera techniques, providing two-dimensional information using transmission, reflection, and dark field imaging of the printed or coated films, and revealing detail on film thickness variations, registration, and particle detection. These techniques are non-contact techniques and apply to individual layers during manufacture. Methods such as light beam induced current mapping, dark lock-in thermographic imaging, electroluminescense imaging, and photoluminescence imaging are being used successfully today in R2R manufacturing of solar cell materials. [48]

  DOE's Manufacturing Demonstration Facility (MDF), established at ORNL, provides unique capabilities to assist industry in adopting new manufacturing technologies to reduce life-cycle energy and greenhouse gas emissions, lower production cost and create new products and opportunities for high paying jobs. [49] The MDF can be a tremendous asset in addressing the above risks and uncertainties.

- **Proprietary Information and Intellectual Property** – Successful implementation of R2R processes within the manufacturing industry will require information exchange, resource partnering, open discussion of ideas, discoveries, and best practices. Key challenges exist in providing an open forum for networking while protecting the proprietary information and intellectual property of the community

- **Technology Characteristics That Impact Policy**

The President's 2012 *National Strategic Plan for Advanced Manufacturing* emphasized the need for increased R&D on advanced materials and innovative manufacturing technologies that have the potential to reduce U.S. manufacturing energy use while enhancing product quality and shortening design cycle times. [50] DOE is responsible for executing programs resulting in the development of competitive new manufacturing processes for U.S. industry to provide state-of-the-art technologies in advanced vehicles, biofuels, solar energy and other clean energy technologies. Technologies areas, such as advanced lightweight materials, membranes and TFTs, have an immediate use for improving products for clean energy applications and are appropriate for manufacturing using R2R processes.

4. Sidebars: Case Studies

4.1. Thin-Film Solar Cell Efficiency Record Set By First Solar (Again) [51] [52]

Figure 17 - First Solar, Inc. Solar Cell Array

Working with DOE National Renewable Energy Laboratory (NREL), First Solar, Inc. set a world record in 2013 for CdTe PV solar cell conversion efficiency, achieving 20.4 percent conversion efficiency. The U.S.-based company recently announced that a cell manufactured at its manufacturing factory and R&D center achieved an efficiency of 21%, the highest on record by a non-concentrating cadmium-telluride (CdTe) cell. Improvement in CdTe PV performance was demonstrated at a rate that dramatically outstrips the trajectory of conventional multicrystalline silicon technologies, which have already plateaued near their ultimate capabilities. First Solar, Inc. has also gone a notch up on multi-crystalline silicon cells, whose efficiency peaked at 20.4% in 2004.

The encouraging fact about the cell is that it has been constructed using processes, such as roll-to-roll, and materials designed for commercial-scale manufacturing, thus making is possibly easier for First Solar to quickly switch to the cell's mass production.

First Solar, Inc. also synergy realized a synergy by partnering with GE Global Research in 2013 with consistent and strong investments in R&D. The advanced technologies and processes developed for the CdTe PV solar cell are already being commercialized and will positively impact performance of future production solar cell modules and power plants.

First Solar has continued to transfer success in R&D into commercial modules, increasing its average production module efficiency to 13.4 percent in the fourth quarter of 2013, up 0.6 percent from 12.9 percent in the fourth quarter of 2012. The company's lead line was producing modules with 13.9 percent average efficiency at the end of 2013. [51]

4.2. Commercial Buildings Integration of Energy Saving Window Coatings [52] [53]


The DOE Building Technologies Office (BTO) works with the commercial building industry to accelerate the uptake of energy efficiency technologies and techniques in both existing and new commercial buildings. By developing, demonstrating, and deploying cost-effective solutions, BTO strives to reduce energy consumption across the commercial building sector by at least 1,600 TBtu. [52] The BTO has several projects in R&D for electrochromic windows, high-insulating windows, and nano-lens window coatings for daylighting and low-e storm windows adoption. [53] R2R manufacturing is used for products like 3M™ window films block up to 60% of the sun’s heat. Transparent, rather than dark or shiny, costs of cooling are saved without sacrificing passive lighting or views. [54]

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